## Pseudo diamagnetism of four component exciton condensates

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(Dated: March 23, 2011)

We analyze the spin structure of the ground state of four-component exciton condensates in coupled quantum wells as a function of spin-dependent interactions and applied magnetic field. The four components correspond to the degenerate exciton states characterized by  $\pm 2$  and  $\pm 1$  spin projections to the axis of the structure. We show that in a wide range of parameters, the chemical potential of the system increases as a function of magnetic field, which manifests a pseudo-diamagnetism of the system. The transitions to polarized two- and one-component condensates can be of the first-order in this case. The predicted effects are caused by energy conserving mixing of  $\pm 2$  and  $\pm 1$  excitons.

PACS numbers: 78.67.-n, 71.36.+c, 42.25.Kb, 42.55.Sa

Introduction.—Cold exciton gases formed by indirect excitons in coupled quantum wells (CQW) represent a solid state bosonic system very rich in new quantum coherent phenomena [1]. Indirect excitons are semiconductor crystal excitations with unique properties: they have long lifetime and spin-relaxation time, can cool down to the temperatures well below the temperature of quantum degeneracy, can travel over large distances before recombination, and can be in situ controlled by applied voltage [2–6]. The indirect excitons form a model system both for the studies of fundamental properties of light and matter and for the development of conceptually new optoelectronic devices [7]. Being formed by heavy holes and electrons, the indirect excitons may have four allowed spin projections to the axis normal to quantum well plane: -2, -1, +1, +2 [8]. The excitons with spin projections -1 and +1 can be coupled to light. Recombining, they emit left- and right-circularly polarized photons, respectively. The exciton states with spin projections -2 and +2 correspond to the optical transitions forbidden in the dipole approximation. These states are usually referred to as "dark excitons". In the absence of external magnetic field dark and bright states of indirect excitons are nearly degenerate. Thus, the gas of indirect excitons can be treated as a four-component degenerate Bose gas. Very interestingly, two components of this gas can be directly studied by the photoluminescence spectroscopy, while two other components are hidden from the observer. Moreover, due to the interplay between dipole-dipole repulsion of excitons and spin dependent exchange interactions of particles, the interactions of indirect excitons are strongly sensitive to their spin and polarization. These features make the gases of indirect excitons a unique system whose thermodynamical properties are expected to be quite different from the properties of multicomponent superfluids [9] and atomic condensates [10] studied before. In this Letter we analyze theoret-

ically the spin and polarization state of Bose-Einstein condensates of indirect excitons (XBEC) at zero temperature. We account for all exciton interaction terms allowed by symmetry and study how the polarization of XBEC changes as a function of the interaction constants and applied magnetic field. We predict the diamagnetic shift of the chemical potential of the exciton condensate if a specific condition on interaction constants is fulfilled.

Condensate at zero magnetic field—We characterize XBEC by a four component order parameter  $\psi_m$  with  $m=\pm 1,\pm 2$ . The indices m denote four allowed exciton spin projections on the structure axis. The Hamiltonian density H in the absence of applied magnetic field can be written as  $(\hbar=1)$ 

$$H = \frac{1}{2M} \sum_{m} |\nabla \psi_{m}|^{2} - \mu n$$

$$+ \frac{1}{2} \sum_{m,m'} V_{m,m'} |\psi_{m}|^{2} |\psi_{m'}|^{2}$$

$$+ W(\psi_{+2}^{*} \psi_{-2}^{*} \psi_{+1} \psi_{-1} + \psi_{+1}^{*} \psi_{-1}^{*} \psi_{+2} \psi_{-2}). \quad (1)$$

Here  $n=\sum_m |\psi_m|^2$  is the total polariton concentration, M is the exciton mass, and  $V_{m,m'}=V_{m',m}$ . In a general case, exciton-exciton interactions are described by five independent constants which come from the amplitude of spin-independent electron-hole interaction and amplitudes of electron-electron and hole-hole interactions in the triplet (parallel spins) and singlet (antiparallel spins) configurations. In the following, we denote  $V_{m,m}=V_0>0$ , which describes interactions between particles having identical spins and accounts for the dipole-dipole and exchange repulsion. The other parameters are  $V_{+2,+1}=V_{-2,-1}=V_0-V_e,\ V_{+2,-1}=V_{-2,+1}=V_0-V_h,$  and  $V_{+2,-2}=V_{+1,-1}=V_0-V_x,$  where  $V_{e,h,x}$  describe exchange interactions between the particles with opposite spins: electrons, holes, and excitons, respectively. In realistic CQW the dipole-dipole repulsion dominates over

the exchange interaction constants, but the polarization state of exciton condensate is governed by small spin-dependent interaction parameters W and  $V_{x,e,h}$ .

The last term in (1) describes the mixing between dark and bright excitons. It appears due to possibility of scattering of two bright excitons into two dark ones and vice versa [11]. This term is switched off if one or more exciton spin components become empty. On the other hand, if all components are occupied, this term reduces the energy of the condensate independently of the sign of W. This is assured by minimization of H over the phase factor  $(\theta_{+2} + \theta_{-2} - \theta_{+1} - \theta_{-1})$ , where  $\theta_m$  is the phase of the m-th component of the order parameter:  $\psi_m = A_m e^{i\theta_m}$ . In the following we set W > 0.

The ground state of XBEC is defined by the signs of parameters

$$u_0 = n(W + V_x + V_e + V_h)/4,$$
 (2a)

$$u_x = n(W - V_x + V_e + V_h)/4,$$
 (2b)

$$u_e = n(W + V_x - V_e + V_h)/4,$$
 (2c)

$$u_h = n(W + V_x + V_e - V_h)/4,$$
 (2d)

as it is shown in Fig. 1, where we denoted  $u_{\min} = \min\{u_x, u_e, u_h\}$ . Note that it is  $u_{\min}$  that defines which particular two-component condensate (TCC) is realized. For example, if  $V_x > W + V_e + V_h$  and  $u_{\min} = u_x < 0$ , either pure dark or pure bright condensate is formed. If  $V_e > W + V_x + V_h$  and  $u_{\min} = u_e < 0$ , the dark and bright components become circular with the same sign of circular polarization. Finally, for  $u_{\min} = u_h < 0$ , the dark and bright components are also circular, but of the opposite signs.

In what follows, we consider the most interesting and presumably most experimentally relevant case of a four-component condensate (FCC), where all parameters (2)

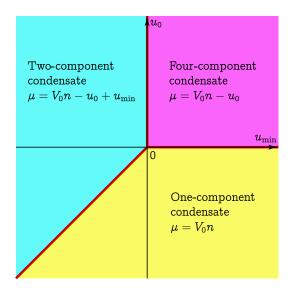


FIG. 1. Showing the state of the exciton condensate for different values of interaction parameters defined in the text.

are positive. The chemical potential in this case is

$$\mu = \mu_0 = V_0 n - u_0. \tag{3}$$

The excitation spectrum of four-component condensate can be found in the usual way [12] by linearizing the Gross-Pitaevskii equation generated by Hamiltonian (1) with respect to small plane-wave perturbation. The spectrum consists in four branches. Three branches have the Bogoliubov dispersion,  $\varepsilon = \sqrt{\varepsilon_0(k)[2u + \varepsilon_0(k)]}$ , where  $\varepsilon_0(k) = k^2/2M$  and u takes the values of  $\mu_0$ ,  $u_e$ , and  $u_h$ . The fourth branch is gaped in k=0 and the quasiparticle energy is

$$\varepsilon_x(k) = \sqrt{[Wn + \varepsilon_0(k)][2u_x + \varepsilon_0(k)]}.$$
 (4)

The gap  $\Delta = \sqrt{2Wnu_x}$  appears due to the mixing term of Hamiltonian (1). We note that its physical origin is the same as of the gap in BCS superconductors: namely, the pare scattering.

Effects of Zeeman splitting—Weak applied magnetic fields affect the exciton condensate mainly due to addition of the Zeeman splitting term  $H_Z$  to the Hamiltonian,

$$H_Z = -\frac{1}{2}(\omega_1 s_1 + \omega_2 s_2),\tag{5}$$

where  $\omega_{1,2} = g_{1,2}\mu_B B$  are the Zeeman splitting energies for single excitons and we introduced the pseudospins of the components  $s_1 = |\psi_{+1}|^2 - |\psi_{-1}|^2$  and  $s_2 = |\psi_{+2}|^2 - |\psi_{-2}|^2$ . The g-factors of dark and bright excitons are  $g_1 = g_h - g_e$  and  $g_2 = g_h + g_e$ , where  $g_e$  and  $g_h$  are g-factors of electrons and heavy-holes, respectively. Both the values and the signs of g-factors can be different as they depend substantially on the quantum well widths [13, 14]. Here and further we neglect the magnetic field effect on internal orbital motion of electrons and holes in the exciton which has been extensively studied in the past [15–18]. That orbital effect is independent on spin and results in the exciton diamagnetic shift similar to the blue shift of atomic lines due to the Langevin diamagnetism.

As we show below, the non-trivial behavior of the equilibrium state of the condensate in magnetic field is due to the possibility of resonant scattering of dark to bright excitons and vice versa described by the last term in the Hamiltonian (1). Minimization of  $H+H_Z$  over  $\psi_m^*$  yields a relation between the amplitudes of the spin components

$$(V_x A_{-m}^2 + V_e A_l^2 + V_h A_{-l}^2) A_m + W A_{-m} A_l A_{-l}$$

$$= \left(V_0 n - \mu - \frac{1}{2} \omega_m\right) A_m, \quad (6)$$

where  $l=\pm 2,\pm 1$  and  $\omega_m=\pm \omega_{1,2}$  for  $m=\pm 1,\pm 2$ , respectively. There are solutions to Eqs. (6) describing FCC, two-component (TCC) and one-component condensate (OCC). We note that there is no solution with three components—the equation corresponding to the single empty component cannot be satisfied due to the

W-term in (6). Physically, it becomes clear if we remember that each exciton is composed by an electron in +1/2 or -1/2 state and a hole in +3/2 or -3/2 state. As soon as the electron (or hole) upper spin component is emptied, the transition from FCC to TCC takes place. If both electrons and holes are fully spin-polarized we obtain OCC.

To describe the polarization of FCC we introduce auxilliary variables

$$t \equiv t_1 = \frac{A_{+1}A_{-1}}{A_{+2}A_{-2}}, \quad t^{-1} \equiv t_2 = \frac{A_{+2}A_{-2}}{A_{+1}A_{-1}},$$
 (7)

that make it possible to write the concentrations,  $n_1 = A_{+1}^2 + A_{-1}^2$  and  $n_2 = A_{+2}^2 + A_{-2}^2$ , and pseudospins of the bright and dark components as

$$n_{1,2} = \frac{Wt_{1,2} + V_x - V_e - V_h}{W(t_1 + t_2) + 2(V_x - V_e - V_h)} n,$$
 (8)

$$s_{1,2} = \frac{(Wt_{1,2} + V_x)\omega_{1,2} + (V_e - V_h)\omega_{2,1}}{(Wt_1 + V_x)(Wt_2 + V_x) - (V_e - V_h)^2}n.$$
(9)

Substitution of these expressions back to definition (7) allows to link t and applied magnetic field by the equation

$$s_1^2 - t^2 s_2^2 = n_1^2 - t^2 n_2^2. (10)$$

Since  $s_{1,2} \propto B$  this equation directly expresses magnetic field as a function of t. Finally, the change of the chemical potential of FCC in magnetic field is given by

$$\mu - \mu_0 = \frac{n}{4} \frac{W(W + V_e + V_h - V_x)(t-1)^2}{W(t+1)^2 - 2t(W + V_e + V_h - V_x)}.$$
 (11)

The system (6) can be easily solved for TCC and OCC. The signs of g-factors of bright and dark excitons define which components remain occupied in high magnetic fields. In what follows, we consider the case  $g_1 \geq g_2 \geq 0$ , when applied magnetic field favors the formation of right-circular condensates [19]. Then, for TCC the amplitudes of components are

$$A_{+1,2}^2 = \frac{1}{2}n + \frac{\omega_{1,2} - \omega_{2,1}}{4V_e}, \quad A_{-1,2}^2 = 0,$$
 (12)

and the chemical potential is

$$\mu - \mu_0 = \frac{n}{4}(W + V_x - V_e + V_h) - \frac{1}{4}(\omega_1 + \omega_2).$$
 (13)

In the region of very strong magnetic fields, the condensate becomes one-component,  $A_{+1} = \sqrt{n}$ ,  $A_{-1} = A_{\pm 2} = 0$ . The chemical potential of OCC is

$$\mu - \mu_0 = \frac{n}{4}(W + V_x + V_e + V_h) - \frac{1}{2}\omega_1.$$
 (14)

The chemical potential of FCC is independent of magnetic field in the case of equal g-factors  $g_1 = g_2$ , since the

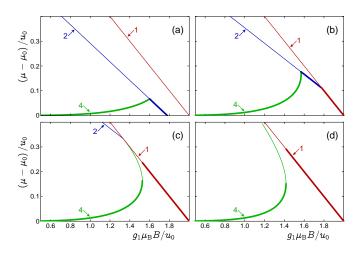


FIG. 2. The magnetic-field dependencies of chemical potentials of four-component (green lines), two-component (blue lines), and one-component (red lines) exciton condensates. (the curves are also labeled by the number of components). Stable and metastable states of a uniform condensate are indicated by bold and thin parts of the curves, respectively. The interaction parameters are: (a)  $W=V_x,\,V_e=V_h=0.5V_x,\,g_2/g_1=0.5$ ; (b)  $W=V_x,\,V_e=V_h=0.5V_x,\,g_2/g_1=0.25$ ; (c)  $W=V_x,\,V_e=(3/8)V_x,\,V_h=(5/8)V_h,\,g_2/g_1=0.25$ ; (d)  $W=3V_x,\,V_e=V_h=0.5V_x,\,g_2/g_1=0.25$ .

bright and dark excitons are polarized in the same way and t=1. However, the behavior of  $\mu$  becomes nontrivial if  $g_1 \neq g_2$ . In this case, if  $W+V_x>V_e+V_h$ , the chemical potential increases with magnetic field as one can see from (11). From the experimental point of it leads to a blue-shift of the emission line, i.e., to an apparent diamagnetic effect. Unlike the conventional diamagnetism, this effect is only related to spin interactions and does not depend on the orbital motion of electrons and holes. It is specific to four-component exciton condensates and does not take place in two-component exciton-polariton condensates [20, 21], where the chemical potential remains independent of the magnetic field up to some critical field.

The effect we discuss can be referred to as pseudo-diamagnetism, since it is not related to the increase of the total energy of the system. In fact, the Zeeman energy (5) decreases quadratically with magnetic field, because  $s_{1,2} \propto B$  for small B [see (9)]. On the other hand, the chemical potential increases  $\propto B^4$  for small B. Clearly, for weak fields this increase can be neglected compared to the usual, orbital diamagnetism  $\propto B^2$ . However, as we show below, the additional pseudo-diamagnetic shift becomes dominant when the bare Zeeman energy approaches the energy of exchange interactions in the exciton condensate and it can result in dramatic changes in the polarization state of the system.

The behavior of chemical potentials (11), (13), and (14) as functions of applied magnetic field is shown in Fig. 2(a-d). For a small difference of g-factors the changes of the ground state of the condensate are continuous,

as it is shown in Fig. 2(a,b). The chemical potential of FCC slightly increases, and at some magnetic field it reaches the chemical potential of TCC. At this point the FCC transforms into TCC: the components  $A_{-2}$  and  $A_{-1}$  vanish. Subsequently, the TCC is transformed into OCC at the higher magnetic field, as it is shown in Fig. 2(b). Note also, that FCC can transform directly into OCC in some range of parameters.

Very interestingly, in a wide range of parameters, namely, for a sufficiently large amplitude of the mixing term W and strong difference of g-factors, the change of the ground state of the condensate is discontinuous [see Fig. 2(c,d)]. In these cases, for a given magnetic field there are two solutions of Eq. (10) for parameter t corresponding to different polarization states of the condensate. The state with a higher value of  $\mu$  is metastable. The FCC disappears at  $B > B_c$ , where  $B_c$  is defined by  $d\mu/dB = \infty$ . If B reaches  $B_c$  from below the FCC with finite occupation of all components transforms discontinuously into OCC. The chemical potential jumps by a finite value in this case.

The discontinuous change of the chemical potential is characteristic for a phase transition of the first-order. In this case one can expect formation of a mixed state of the system for B close to  $B_c$ , where FCC and OCC with different concentrations of excitons coexist, similarly to how it happens in the case of vapor-liquid phase transition.

In conclusion, we have shown that applied magnetic field suppresses the mixing of dark and bight excitons and leads to pseudo-diamagnetic increase of the chemical potential of exciton condensate, provided the dark and bright excitons possess different g-factors. The interplay between spin-dependent exciton-exciton interactions and Zeeman effect can lead to the first order transition between four-component and one- or two-component condensates.

We are grateful to B. L. Altshuler and M. M. Glazov for valuable discussions. This work was supported in part by DGAPA-UNAM under Project No. IN112310 and by the EU FP7 IRSES Project POLAPHEN.

- [2] L. V. Butov, L. S. Levitov, A. V. Mintsev, B. D. Simons, A.C. Gossard, and D.S. Chemla, Phys. Rev. Lett. 92, 117404 (2004).
- [3] R. Rapaport, G. Chen, D. Snoke, S. H. Simon, L. Pfeiffer, K. West, Y. Liu, and S. Denev, Phys. Rev. Lett. 92, 117405 (2004).
- [4] G. Chen, R. Rapaport, S. H. Simon, L. Pfeiffer, and K. West, Phys. Rev. B 71, 041301 (2005).
- [5] M. Haque, Phys. Rev. E 73, 066207 (2006).
- [6] Sen Yang, L. V. Butov, L. S. Levitov, B. D. Simons, and A. C. Gossard, Phys. Rev. B 81, 115320 (2010).
- [7] Y. Y. Kuznetsova, M. Remeika, A. A. High, A. T. Hammack, L. V. Butov, M. Hanson, and A. C. Gossard, Optics Letters 35, 1587 (2010).
- [8] J. R. Leonard, Y. Y. Kuznetsova, Sen Yang, L. V. Butov, T. Ostatnicky, A. Kavokin, and A. C. Gossard, Nano Letters 9, 4204 (2009).
- [9] M. M. Salomaa and G. E. Volovik, Rev. Mod. Phys. 59, 533 (1987).
- [10] M. Ueda and Y. Kawaguchi, arXiv:1001.2072.
- [11] C. Ciuti, V. Savona, C. Piermarocchi, A. Quattropani, and P. Schwendimann, Phys. Rev. B 58, 7926 (1998).
- [12] See, e.g., E. M. Lifshitz and L. P. Pitaevskii, Statistical Physics, part 2 (Pergamon Press, New York, 1980).
- [13] M. J. Snelling, E. Blackwood, C. J. McDonagh, R. T. Harley, and C. T. B. Foxon, Phys. Rev. B 45, 3922 (1992).
- 14] M. Bayer et al., Phys. Rev. B 65, 195315 (2002).
- [15] R. J. Elliott and R. Loudon, J. Phys. Chem. Solids 15, 196 (1960).
- [16] L. P. Gor'kov and I. E. Dzyaloshinskii, Zh. Eksp. Teor. Fiz. 53, 717 (1967) [Sov. Phys. JETP 26, 449 (1968)].
- [17] I. V. Lerner and Yu. E. Lozovik, Zh. Eksp. Teor. Fiz. 78, 1167 (1980) [Sov. Phys. JETP 51, 588 (1980)].
- [18] Yu. M. Lozovik, I. V. Ovchinnikov, S. Yu. Volkov, L. V. Butov, and D. S. Chemla, Phys. Rev. B 65, 235304 (2002).
- [19] The other cases can be reproduced by simple relabeling of the components and interaction constants, since Eqs. (6) are invariant under simultaneous transformations  $V_e \leftrightarrow V_h$ ,  $A_{+2} \leftrightarrow A_{-2}$ , and  $g_2 \to -g_2$ .
- [20] Yu. G. Rubo, A. V. Kavokin, and I. A. Shelykh, Phys. Lett. A 358, 227 (2006).
- [21] A. V. Larionov, V. D. Kulakovskii, S. Höfling, C. Schneider, L. Worschech, and A. Forchel, Phys. Rev. Lett. 105, 256401 (2010).

<sup>[1]</sup> L. V. Butov, A. C. Gossard, and D. S. Chemla, Nature 418, 751 (2002).